

## Neutron Scattering

**Description:** The principal objective of this program is the study of cooperative phenomena in complex solids by elastic and inelastic neutron scattering. Aspects investigated include structural and magnetic phase transformations, magnetic structure, and elementary excitations such as spin-waves and phonons. Neutron scattering provides a unique tool for the study of these phenomena, and experiments are carried out at neutron facilities in the U.S. and abroad. The ultimate motivation is the broad understanding of underlying physical principles. In many cases, complementary studies are performed in collaboration with other BNL groups in Condensed Matter Physics and Materials Science. An effort to grow (at BNL) the single-crystal samples necessary for some of these studies has been initiated. There are also efforts, in collaboration with BNL's Center for Neutron Science (CNS) as well as with neutron scientists at other labs and universities, to design and build new spectrometers for the Spallation Neutron Source and the High Flux Isotope Reactor, both at Oak Ridge National Laboratory.

### **Program Highlights:**

- *Optical phonon anomalies in a stripe-ordered nickelate are similar to those found in superconducting cuprates.* A study of bond-stretching phonons in  $\text{La}_{1.69}\text{Sr}_{0.31}\text{NiO}_4$  reveals a 10% softening at the zone boundary but not at the zone center, which is clearly induced by the hole doping. (cond-mat/0107635)
- *Stripes in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  rotate from diagonal to vertical at the superconductor-insulator boundary.* Elastic magnetic diffraction studies indicate that diagonal stripes are present throughout the "spin-glass" regime, from the antiferromagnetic boundary, at  $x = 0.02$ , up to the transition to superconductivity, at  $x \approx 0.055$ . (cond-mat/0101320)
- *Nanometer-sized polarized domains relax or ferroelectrics have a dramatic impact on optical phonons.* The lowest-frequency transverse optical mode is strongly overdamped for wave vectors smaller than a characteristic value. PRL **84**, 5216 (2000).
- *Continuum spin excitations observed convincingly for the first time in an  $S = 1$  chain system.* The measurements were performed on the Haldane-chain compound  $\text{CsNiCl}_3$ . PRL **87**, 017202 (2001).
- *Magnetic-field-induced order in a quantum-disordered system.* Commensurate long-range order was induced by applying a strong magnetic field to the Haldane-gap system  $\text{Ni}(\text{C}_5\text{H}_{14}\text{N}_2)_2\text{N}_3(\text{ClO}_4)_2$ . Europhys. Lett. **55**, 868 (2001).
- *Charge and spin ordering have very different energy scales in  $\text{La}_{1.5}\text{Sr}_{0.5}\text{CoO}_4$ .* Checker-board charge order sets in at 825 K, whereas staggered magnetic order appears only below 30 K. PRL **85**, 4353 (2000).
- New Book: *Neutron Scattering with a Triple-Axis Spectrometer: Basic Techniques*, by G. Shirane, S. M. Shapiro, and J. M. Tranquada, to be published by Cambridge University Press in January 2002. The first guide book to explain how to exploit the power, while avoiding the pitfalls, of neutron scattering with a triple-axis spectrometer.

### **Impact**

- Our experimental studies of charge-stripe correlations in hole-doped antiferromagnets, especially cuprates and nickelates, has stimulated a considerable amount of related theoretical and experimental work, as reflected in citations.
- The identification of a monoclinic phase of  $\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$  (PZT) at the composition corresponding to optimal piezoelectric performance has allowed a complete theoretical understanding of this important class of materials, and has led to predictions of improved performance in materials with special nanoscale ordering of the component elements.

### **Interactions:**

- BNL: Condensed Matter Physics groups, Materials Science groups
- U.S. Universities: MIT, U. Delaware, U. Pennsylvania, U. Connecticut, Johns Hopkins U., Rutgers U., Iowa State U.
- Non-U.S. Universities: U. Tokyo, Tohoku U., Kyoto U., U. Cologne, Oxford U., T.U. Munich, U. Toronto, Nagoya U.
- U.S. National Laboratories: Ames Lab., NIST, ORNL, LANL
- Non-U.S. Laboratories: CEA/Grenoble, HASYLAB, HMI, ILL, ISSP, JAERI, Karlsruhe, LLB, PSI, RAL, RIKEN
- Industrial: Lucent Technologies, CRIEPI (Japan)

### **Personnel (current):**

J. M. Tranquada (group leader), G. Gu, S. M. Shapiro, G. Shirane, I. Zaliznyak; Post-docs: M. Hücker, H. Woo

### **Recognition:**

- 1 member of the NAS and former recipient of the Buckley prize; 3 Fellows of the APS
- 1 Presidential Early Career/DOE Young Investigator Award
- 29 invited talks at national and international conferences in last 3 years

**Annual Budget:** \$1710K

## **Neutron Scattering**

### **Scientific Staff:**

<b>Genda Gu</b> (arrived 2/01)	Single-crystal growth, especially complex oxides
<b>Stephen M. Shapiro</b> (also head of CNS)	Martensitic transformations, spin glasses, <i>f</i> -electron systems
<b>Gen Shirane</b>	Phase transitions, magnetism, superconductivity
<b>John M. Tranquada</b>	Superconductivity, antiferromagnetism, charge order in complex oxides
<b>Igor Zaliznyak</b> (arrived 6/99)	Quantum magnetism; charge order in complex oxides
<b>Andrey Zheludev</b> (departed 8/01)	Quantum magnetism in low-dimensional systems

### **Post Docs:**

<b>Markus Hücker</b> (arrived 4/01)	Magnetism and structural transitions in cuprates and nickelates
<b>Hyungje Woo</b> (arrived 9/01)	Colossal magnetoresistance; to be stationed at ISIS

### **Support Staff:**

2 Technicians

1/2 Secretary

### **Sources of Funding beyond DOE:**

- \$140K/yr U.S.-Japan Cooperative Research Program on Neutron Scattering

### **Facilities:**

- 13-T vertical-field split-coil magnet with <sup>3</sup>He cryostat insert; currently being installed at NIST Center for Neutron Research
- 1800°C furnace designed for neutron scattering experiments
- NEC infrared image furnace (SC-I-MDH-20020-SP), for growth of single crystals by the travelling-solvent floating-zone technique, to be installed 11/01
- U.S.-Japan triple-axis spectrometer to be installed at C(old)G(uide)-4 in the guide hall at HFIR by 2003, in collaboration with BNL's Center for Neutron Science (CNS)
- Scientific collaboration with the neutron scattering group at CEA/Grenoble (led by L. P. Regnault) provides access to Cooperative Research Group instruments at the ILL (Instruments: IN22, IN12, D23)
- Part of the H7 triple-axis spectrometer was installed on spectrometer BT-9 at the NIST Center for Neutron Research (NCNR), through BNL's CNS, improving that instrument and providing preferential access
- Post-doc stationed at ISIS should enhance access to state-of-the-art time-of-flight instruments such as MAPS

### **Future Directions:**

- Will improve capabilities for materials synthesis and characterization
- In concert with BNL's Center for Neutron Science, we are pursuing the development of a hybrid spectrometer (HYSPEC) for the study of elastic and inelastic scattering from single crystals, to be built at SNS
- Through CNS, intend to establish a scientific alliance with NCNR, involving a new staff scientist stationed at NCNR
- Will participate, with the CNS, in development of new state-of-the-art triple-axis spectrometer on C(old)G(uide)-1 at HFIR, in collaboration with ORNL and university researchers

## **Neutron Scattering**

### **Major Outside Collaborators:**

President R. J. Birgeneau, *University of Toronto*  
Prof. P. Böni, *Technical University of Munich, Germany*  
Prof. A. Boothroyd, *Oxford University, England*  
Dr. P. Bourges, *Laboratoire Léon Brillouin, France*  
Prof. M. Braden, *University of Cologne, Germany*  
Prof. C. Broholm, *Johns Hopkins University*  
Prof. D. J. Buttrey, *University of Delaware*  
Prof. P. Canfield, *Iowa State University*  
Dr. S.-W. Cheong, *Rutgers University*  
Prof. Y. Endoh, *Tohoku University, Japan*  
Prof. Y. Fujii, *ISSP, University of Tokyo, Japan*  
Dr. M. Fujita, *Kyoto University, Japan*  
Dr. P. M. Gehring, *National Institute of Standards and Technology*  
Prof. K. Hirota, *Tohoku University, Japan*  
Prof. K. Katsumata, *RIKEN, Japan*  
Dr. S.-H. Lee, *National Institute of Standards and Technology*  
Prof. Y. S. Lee, *Massachusetts Institute of Technology*  
Dr. T. Lograsso, *Ames Laboratory*  
Dr. M. Matsuda, *JAERI, Japan*  
Dr. Y. Moritomo, *Nagoya University, Japan*  
Dr. S. Nagler, *Oak Ridge National Laboratory*  
Dr. T. Perring, *ISIS, Rutherford-Appleton Laboratory, United Kingdom*  
Dr. L. Pintschovius, *Forschungszentrum Karlsruhe, Germany*  
Dr. L. P. Regnault, *CEA/Grenoble, France*  
Prof. J. Schneider, *HASYLAB, Germany*  
Prof. C. Stassis, *Iowa State University*  
Prof. S. Uchida, *University of Tokyo, Japan*  
Prof. K. Uchinokura, *University of Tokyo, Japan*  
Dr. S. Wakimoto, *University of Toronto*  
Prof. K. Yamada, *Kyoto University, Japan*

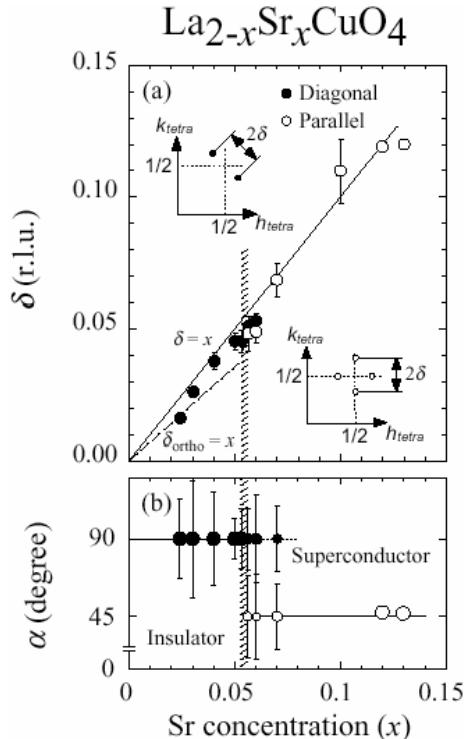
### **Examples of Internal BNL Collaborations**

- *Stripe correlations in  $La_{2-x}Sr_xNiO_4$ :* Tranquada, Hücker, and Woo collaborate with Emery and Tsvelik (Condensed Matter Theory), Homes (Electron Spectroscopy), Vogt (Powder Diffraction), Hill (X-ray Scattering), Li and Moodenbaugh (Superconductivity), Zhu (Electron Diffraction)
- *Studies of superconducting cuprates:* Gu, Shirane, Tranquada, and Hücker collaborate with Emery and Tsvelik (CM Theory), Johnson, Valla, and Homes (Electron Spectroscopy), Hill, Kim, and Nelson (X-ray Scattering), Li and Moodenbaugh (Superconductivity)
- *Structural studies of PZT and related ferroelectrics:* Shirane collaborates with Noheda and Cox (Powder Diffraction), Kim (X-ray Scattering)
- *Giant dielectric response in  $CaCu_3Ti_4O_{12}$ :* Shapiro collaborates with Homes (Electron Spectroscopy), Vogt (Powder Diffraction)
- *Superconductivity in  $MgB_2$ :* Gu collaborates with Li and Moodenbaugh (Superconductivity), Vogt (Powder Diffraction), Zhu (Electron Diffraction)
- *Charge and spin order in cobaltates:* Zaliznyak and Tranquada collaborate with Hill and Kim (X-ray Scattering), Zachar (CM Theory visitor)

## Neutron Scattering

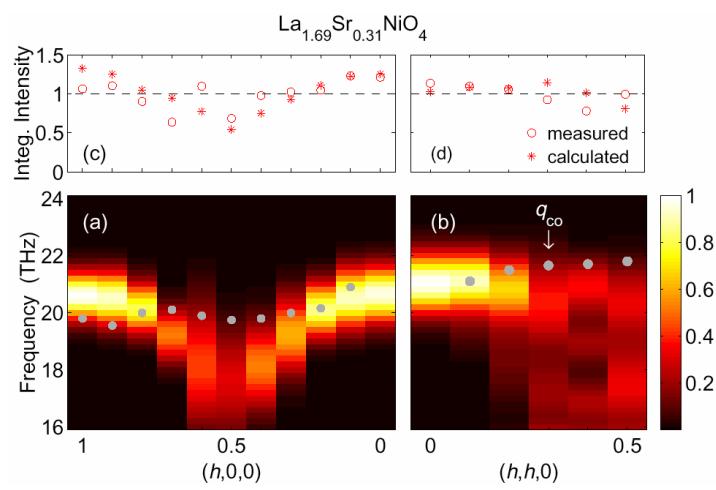
### Highlights:

Charge stripes change orientation at the insulator -superconductor boundary in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$



The superconductivity in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  is obtained by doping holes into an antiferromagnetic insulator. It has been known for over a decade that the antiferromagnetic order disappears at  $x = 0.02$ , while superconductivity first appears at  $x = 0.055$ . Magnetic correlations survive in the superconducting state, but are incommensurate. Are the incommensurate magnetic correlations associated with the response of Fermi-liquid-like conduction electrons, or are they evidence for antiphase antiferromagnetic domains defined by the segregation of the dopant-induced holes into charge stripes? Important evidence supporting the latter case has come from an extensive investigation of the “spin-glass” regime, between the antiferromagnetic and superconducting phases. This work has revealed incommensurate magnetic order throughout the whole regime; however, the magnetic modulation is at 45° to the Cu-O bond direction in the insulating region ( $x < 0.055$ ), rotating to be parallel to the Cu-O bonds in the superconducting phase at larger  $x$ . In the figure at the left, (a) shows the incommensurate splitting as a function of Sr concentration, while (b) shows the angle of the modulation wave vector with respect to the [110] direction. Current work is focusing on the low-temperature changes in magnetic order for  $x < 0.02$  previously identified by NMR and  $\mu\text{SR}$  studies. [G. Shirane (BNL); S. Wakimoto (U. Toronto); M. Fujita, K. Yamada (U. Kyoto); M. Matsuda (JAERI); H. Hiraka (Tohoku U.); P.M. Gehring, S.-H. Lee (NIST)]

Phonon anomalies in stripe-ordered nickelate are similar to those in cuprates



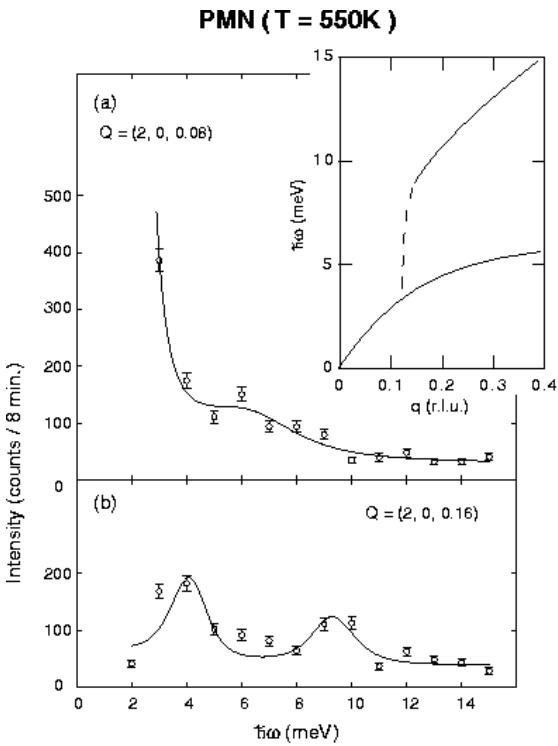
The significance of phonons and electron-phonon interactions in cuprate superconductors has recently seen a revival in interest. If charge stripes are relevant to the cuprates, then screening of the charge modulation by the lattice would be expected. To test this possibility, the bond-stretching phonon modes in a stripe-ordered nickelate model system ( $\text{La}_{1.69}\text{Sr}_{0.31}\text{NiO}_4$ ) were studied by neutron scattering. The results are summarized in the figure on the left (gray symbols in (a) and (b) indicate results for undoped  $\text{La}_2\text{NiO}_4$ ). Panel (a) shows the dispersion along the direction parallel to the Ni-O bonds; the softening between  $h = 0$  and  $h = 0.5$  is very similar to what is observed in cuprates. Panel (b) shows a splitting of frequencies for modes propagating parallel and perpendicular to the charge stripes.

The absence of any special anomaly at the charge ordering wave vector (indicated by the arrow) suggests that collective interactions between the stripes and the lattice, as would occur with conventional charge-density waves, may not be important here. Instead, the electron-phonon interaction may involve individual stripes. New experiments are being initiated to study the related phonon branches in detwinned crystals of the superconductor  $\text{YBa}_2\text{Cu}_3\text{O}_7$ .

[J.M. Tranquada (BNL); K. Nakajima (ISSP, U. Tokyo); M. Braden (U. Cologne); L. Pintschovius (Karlsruhe); R.J. McQueeney (LANL)]

## Nanodomains in ferroelectrics

The lead-oxide class of perovskite relaxor compounds exhibit remarkable piezoelectric properties that far exceed those of conventional  $\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$  (PZT) ceramics, which are the present day material of choice for most actuator and transducer-based device applications. Most researchers view  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ , or PMN as the prototype of this class. PMN is a complex system that differs from simple perovskite  $\text{ABO}_3$  compounds like  $\text{PbTiO}_3$  by virtue of the mixed-valence character of the B-site. Charge neutrality imposes the respective  $\text{Mg}^{2+}$  and  $\text{Nb}^{5+}$  stoichiometry of  $1/3:2/3$ , while moderate doping with ferroelectric  $\text{PbTiO}_3$  enhances the piezoelectric response by more than a factor of two.

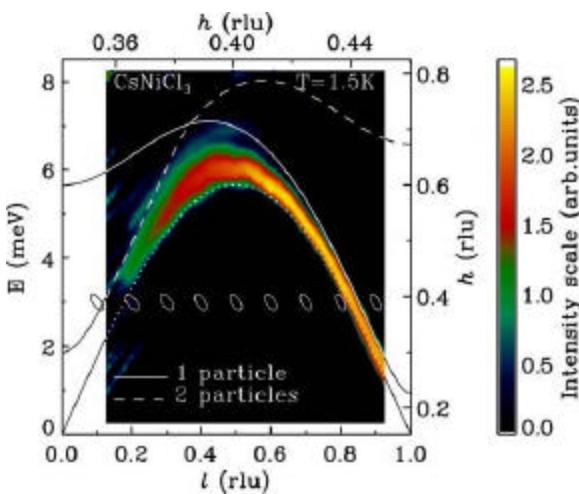


The built-in disorder of the B-site breaks the local translational symmetry, and gives rise to sharply varying electric field gradients. The associated dielectric susceptibility of PMN exhibits a broad or “diffuse” peak at a highly frequency-dependent temperature  $T_{\max} = 230\text{ K}$ , yet no spontaneous polarization develops at any temperature in zero applied field. Perhaps the most seminal feature common to all relaxor systems studied to date is the presence of randomly-oriented regions of local polarization several unit cells in size, known as polar nanoregions (PNR), that only vanish several hundred degrees above  $T_{\max}$ , at a temperature known as the Burns temperature  $T_d$  ( $620\text{ K}$  for PMN).

Our current research uses triple-axis neutron scattering methods to gauge the effects of the PNR on the lattice dynamics in PMN. Of particular interest are the lowest-frequency transverse optic (TO) phonon modes, as the corresponding zone center TO mode is responsible for driving the cubic-to-tetragonal phase transition in  $\text{PbTiO}_3$  at  $763\text{ K}$ . We find that the PNR strongly dampen the TO modes for wavevectors  $q$  less than some characteristic wavevector  $q_{\text{wf}}$ . Figure 1 shows two constant- $Q$  scans measured below and above  $q_{\text{wf}}$  at  $550\text{ K}$ . The damping effectively spreads out the spectral weight of the phonon scattering cross section such that the corresponding TO branch gives the false impression of diving into the TA branch, as shown schematically in the inset ( $q_{\text{wf}} = 0.12\text{ rlu}$ ). This feature is known as the “waterfall” anomaly, and has been observed in many other relaxor systems as well. Its exact relation to the size distribution and density of the PNR is still being studied. [G. Shirane (BNL); P.M. Gehring (NIST); S.E. Park (Penn. State)]

## Continuum excitations in quantum spin liquids

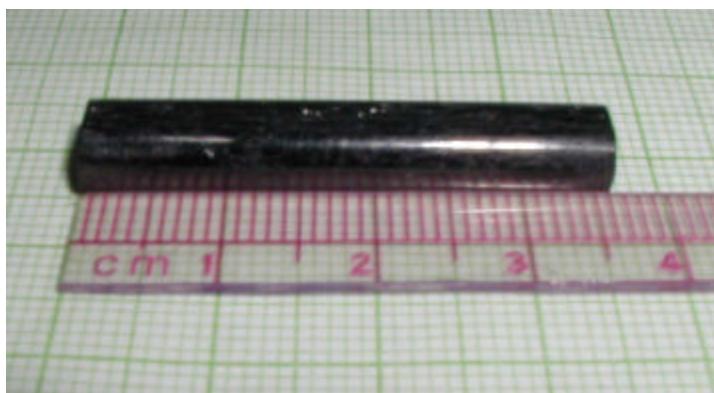
Continuum excitations appear to be a generic feature of quantum liquids in the vicinity of a zero-temperature phase transition. In 1D  $S=1/2$  Heisenberg antiferromagnet (HAFM) the whole triplet spectrum is a continuum made of an even number of  $S=1/2$  freely propagating spinons. In another quantum liquid, superfluid  $^4\text{He}$ , a transformation of the top-of-the-band “maxon” excitation into a broad continuum is observed under pressure, when the roton gap is suppressed and the system is driven towards crystallization.



In recent neutron scattering experiments we studied the low-temperature excitation spectra in two quantum spin liquids, a quasi-1D  $S=1$  Haldane gap HAFM  $\text{CsNiCl}_3$ , and a quantum-disordered 2D HAFM piperazinium hexachlorodocuprate (PHCC). We found that at least in a part of the Brillouin zone both systems do not support coherent  $S=1$  magnon modes, and a continuum band of states is formed instead. In  $\text{CsNiCl}_3$  the single mode approximation fails and a finite energy width appears in the dynamic correlation function  $S(\mathbf{q}, \omega)$  for  $q < 0.6p$ , whose width increases with decreasing  $q$ . This finding is consistently in two different high-luminosity experimental setups, which we have pioneered. In PHCC, with almost isotropic two-dimensional dispersion of a single magnon, cross-over to a continuum occurs close to the top of the dispersion at small  $q$ , where coherent mode “hits” the two-magnon band, so that they “merge” into a single broad feature. These results have the potential to change the way we think of magnetic excitations in this important class of magnets. [I. Zaliznyak (BNL); S.H. Lee (NIST); S.V. Petrov]

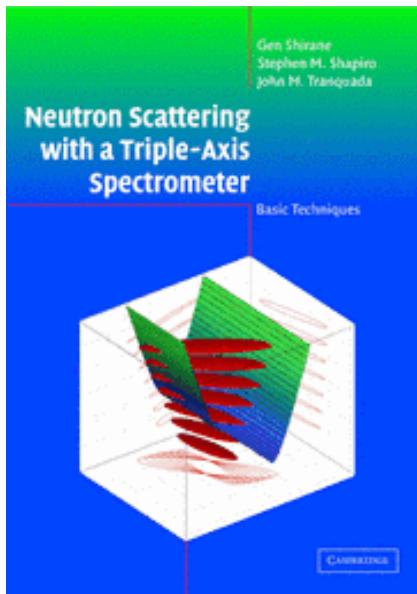
## Growth of new, large cuprate single crystals

While awaiting the arrival of our new furnace from NEC, Genda Gu spent the summer of 2001 at McMaster University, working in the laboratory of Prof. Bruce Gaulin. There he made use of a 4-mirror CSI image furnace to grow



large single crystals of the little-studied bilayer cuprate  $\text{La}_{1.88}\text{Ca}_{1.12}\text{Cu}_2\text{O}_6$ , such as the one shown in the figure. (In exchange for use of the furnace, Genda transferred knowledge on how to grow large crystals of superconducting  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+ \delta}$ ) Initial measurements at NCNR on a smaller crystal demonstrated a modest mosaic width of  $0.3^\circ$ . The as-grown crystals are hole-doped (0.06 holes per Cu) but non-superconducting; a broad superconducting transition at about 30 K was achieved in one piece of crystal by annealing in high-pressure oxygen. The nature of the magnetic correlations in the as-grown crystals will be studied in the near future, when HFIR and NCNR start up again.

## Neutron Scattering with a Triple-Axis Spectrometer: Basic Techniques



G. Shirane, S. M. Shapiro, and J. M. Tranquada have written a book that documents the knowledge base developed from thirty years of experience at the High Flux Beam Reactor. Introductory chapters summarize useful scattering formulas and describe the components of a spectrometer, followed by a comprehensive discussion of the resolution function and focusing effects. Later sections include simple examples of phonon and magnon measurements, and an analysis of spurious effects in both inelastic and elastic measurements, and how to avoid them. Finally, polarization analysis techniques and their applications are covered. This guide will allow graduate students and experienced researchers new to neutron scattering to make the most efficient use of their experimental time.

<http://uk.cambridge.org/physics/catalogue/0521411262/>

## Neutron Scattering

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